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Modern Solid State Laser Materials

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**Lawrence
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Modern Solid State Laser Materials

by

William F. Krupke

This document contains visual aids used in an invited talk entitled "Modern Solid State Laser Materials," presented at the Conference on Lasers and Electro-Optics (CLEO) held in Anaheim, California, on June 20, 1984. Collaborators in the work presented include Howard Guggenheim of AT&T Bell Laboratories, Murray Hill, New Jersey; Frank Bruni of Materials Progress Corporation, Santa Rosa, California; and colleagues at Lawrence Livermore National Laboratory (LLNL): John Caird, Herbert Newkirk, Michael Shinn, Stanley Stokowski, and Ray Wilder. Interest at LLNL in solid state lasers focuses on evaluating the potential of solid state laser media for high average power applications, including inertial fusion power production. To be successful, solid state laser materials will have to attain higher performance levels of energy-storage density, efficiency, and aperture scaling than heretofore. This talk identifies the relevant bulk material parameters characterizing average power capacity and uses chromium and neodymium co-doped gadolinium scandium gallium garnet (Nd:Cr:GSGG) as an example of a laser material with improved laser properties relative to Nd:YAG (plausible large-scale growth, more efficient spectral coupling to xenon flashlamp radiation, reduced stimulated emission cross section, adequate thermal shock and optical damage threshold parameters, etc.). Recently measured spectroscopic, kinetic, and thermo-mechanical properties of Nd:Cr:GSGG are given.

Tunable lasers based on phonon-terminated transitions of transition-metal doped solids is of current interest with respect to a variety of applications. We have recently observed stimulated emission in the phonon-terminated $^4T_2 + ^4A_2$ band of Cr^{3+} in two fluoride crystalline host materials doped with trivalent chromium: the fluoride garnet $\text{Cr:Li}_3\text{Na}_3\text{Ga}_2\text{F}_{12}$ and the fluoride perovskite Cr:KZnF_3 . To be efficient as a short pulse laser (to enable subsequent nonlinear wavelength shifting operations), one desires a stimulated emission cross

section of a few times 10^{-20} cm². This value will permit the stored energy to be efficiently extracted at a beam fluence below the optical damage fluence. At the same time, one desires an energy storage lifetime of at least 100 μ sec so that flashlamp pumping can be carried out with reasonably high lamp shot life. These requirements are more readily realizable in fluoride based materials than in oxides because of the significantly lower index of refraction of the former compared to the latter. In this respect, the system Cr:KZnF₃ offers an improved set of laser spectroscopic parameters for efficient tunable laser action near 800 nm. While this material (indeed most fluorides) possesses less rugged thermo-mechanical properties compared to many oxide crystals, it may prove to be sufficiently robust for a number of useful applications. Measurement of relevant physical properties are currently in progress.

Formal reports describing the measurements and data presented in this talk are in preparation currently.



Cr:Nd:Gadolinium oxide garnets

- Spectroscopy
- Laser parameters
- Thermo-mechanical properties
- Growth

Cr-tunable laser materials

- Fluoride/oxide garnets
- Fluoride perovskites/elpasolites
- Laser parameters

Seminal references



Cr-Nd Co-doped materials

D. Pruss, et al.

Appl. Phys. B28, 355 (1982).

E.V. Zharikov, et al.

Sov. J. Quantum Electron. 12, 338 (1982).

E.V. Zharikov, et al.

Sov. J. Quantum Electron. 13, 82 (1983).

Cr-doped tunable materials

B. Struve, et al.

Appl. Phys. B28, 235 (1982).

B. Struve, et al.

Appl. Phys. B30, 117 (1983).

E.V. Zharikov, et al.

Sov. J. Quantum Electron. 13, 1274 (1983).

U. Brauch, et al.

Optics Communications 49, 61 (1984).

Modern crystalline lasers – collaborators



AT&T Bell Laboratories

Howard Guggenheim

Materials Progress, Inc.

Frank Bruni

LLNL

John Caird

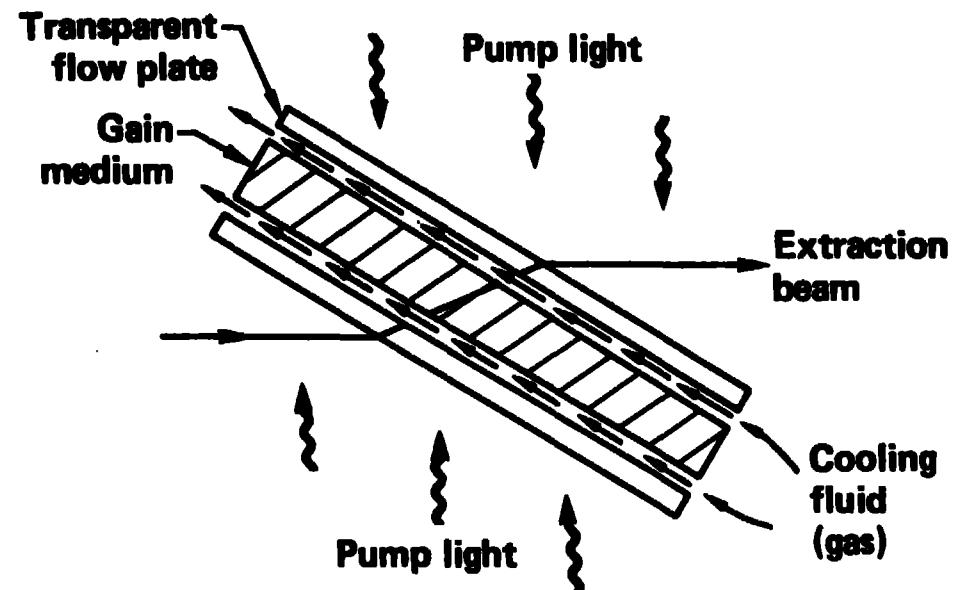
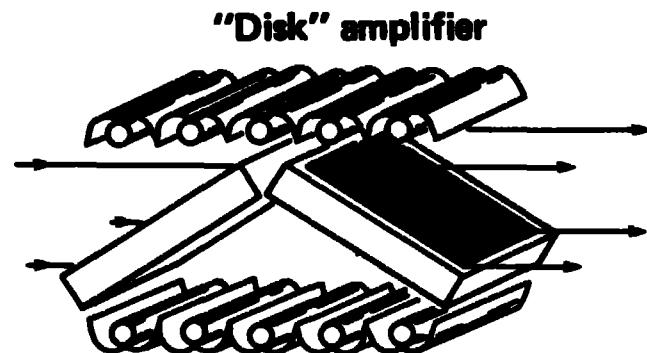
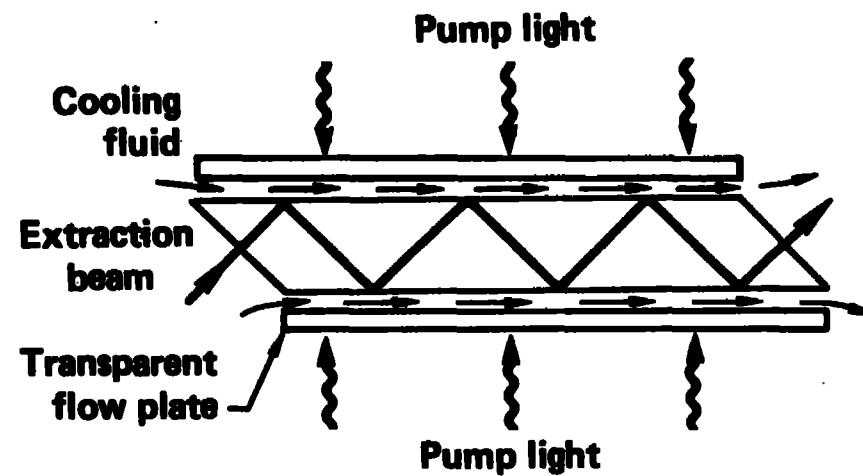
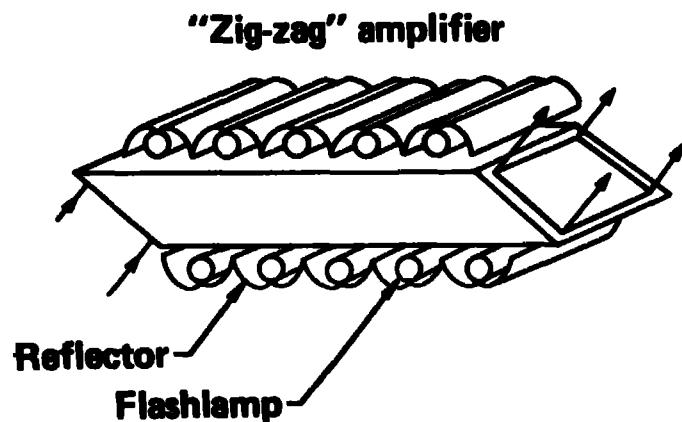
Herb Newkirk

Mike Shinn

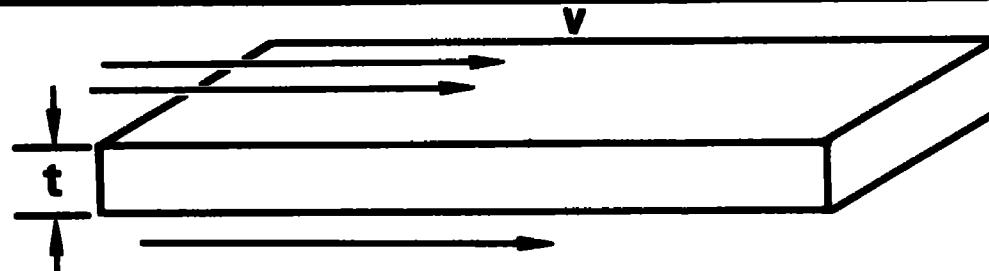
Stan Stokowski

Ray Wilder

High average power laser amplifier geometries



Thermal power capacity of a uniformly-heated flow-cooled plate



$$\frac{\text{Thermal power}}{\text{Unit volume}} = P_v = \frac{12 R_T b}{t^2} \approx (0.5-2) \left\{ \frac{\text{Optical power}}{\text{Unit volume}} \right\}; R_T = \frac{(1-\nu)\kappa S_T}{\alpha E}$$

R_T = Thermal shock parameter

ν = Poisson's ratio

κ = Thermal conductivity

S_T = Tensile yield strength

α = Thermal expansion coefficient

E = Young's modulus

$b = \frac{\text{Thermally induced stress}}{\text{Tensile yield stress}}$

Three high power strategies:

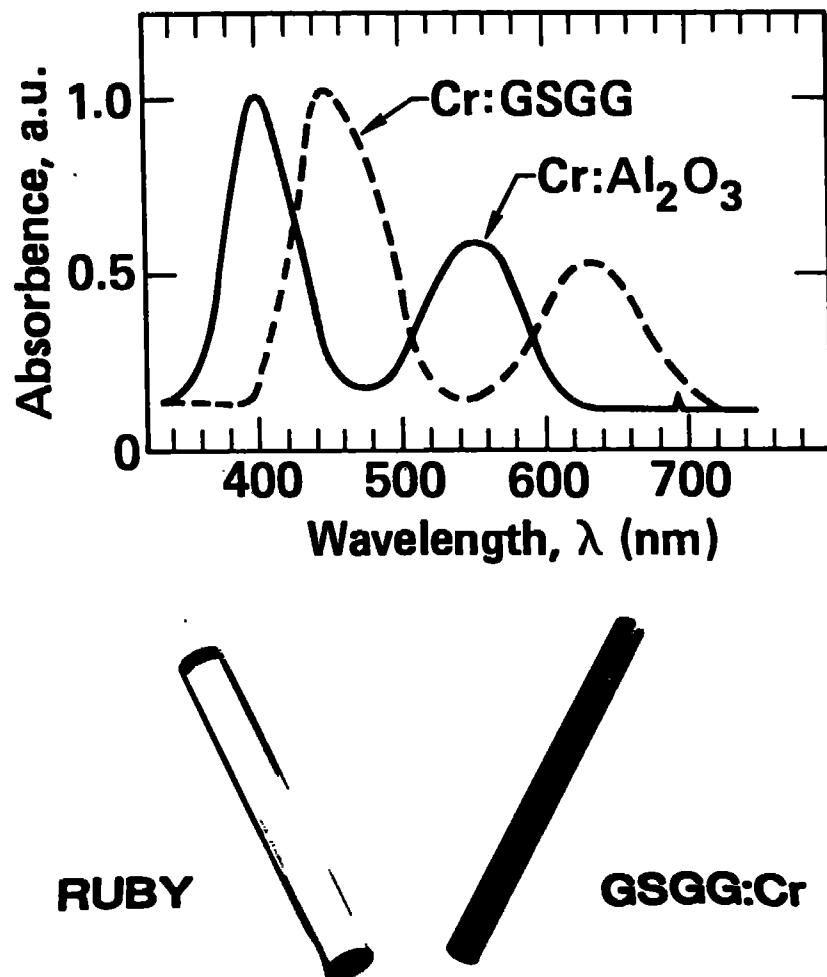
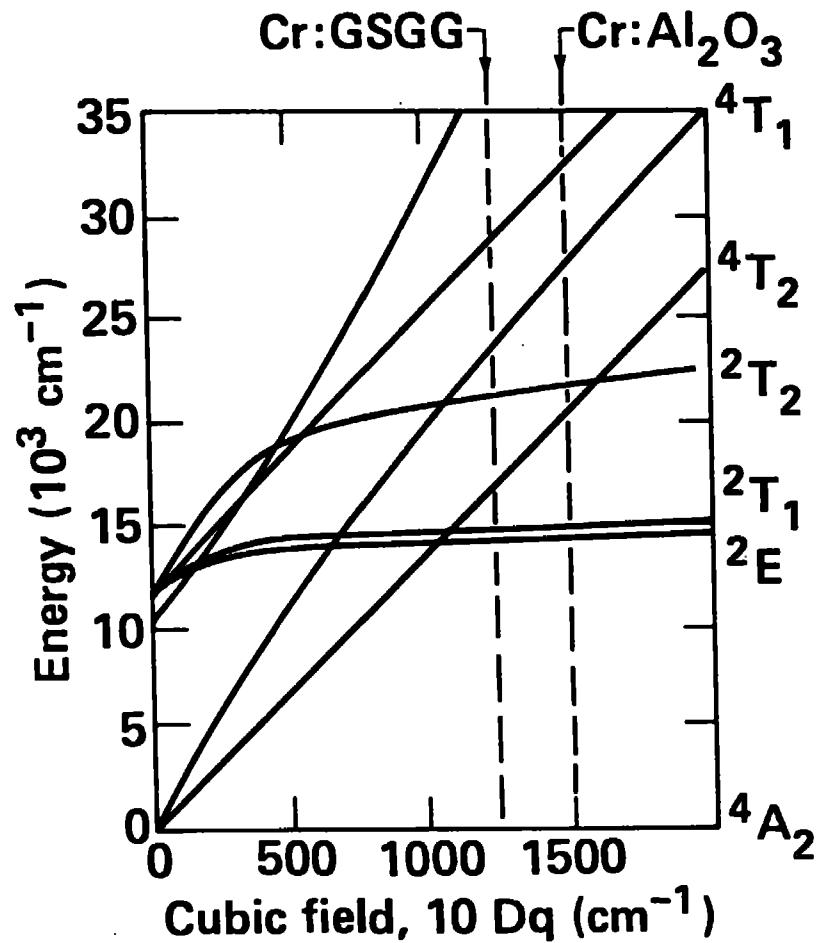
– Materials with high R_T :

- High $\kappa \Rightarrow$ crystals

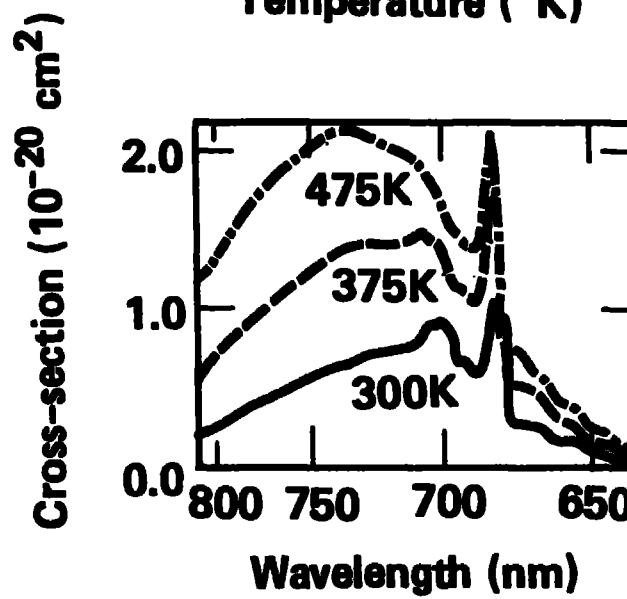
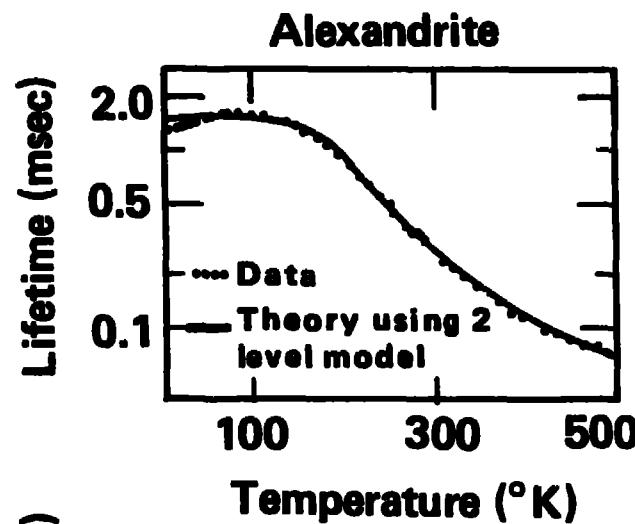
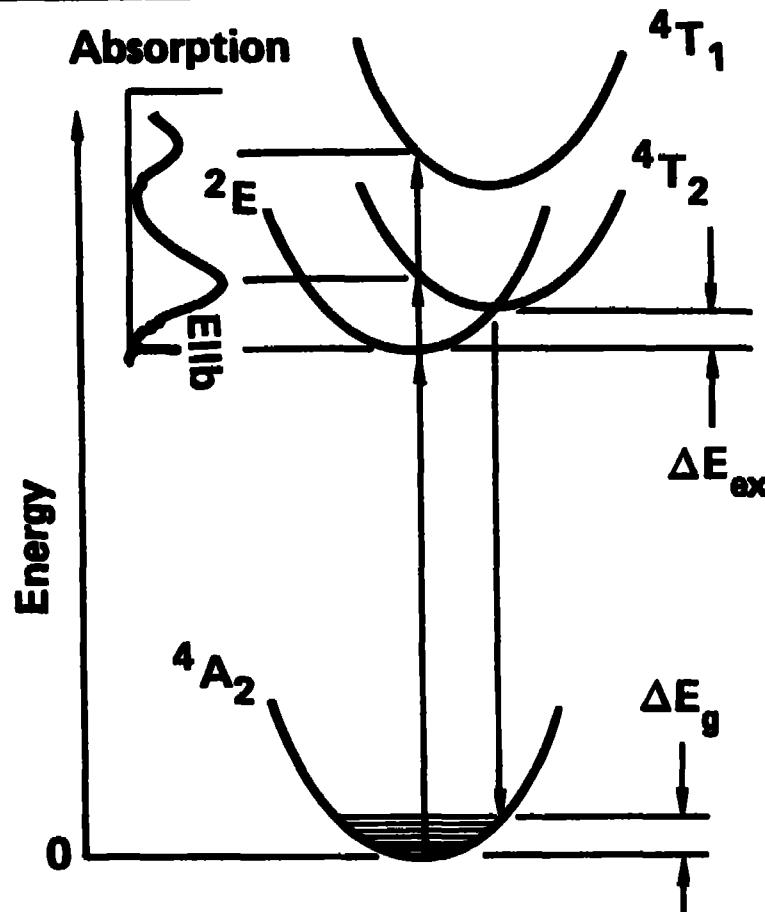
- Low $\alpha \Rightarrow \begin{cases} \text{Low expansion glasses} \\ \text{Glass ceramics} \end{cases}$

– Small $t \Rightarrow$ Composite plate structures

$3d^3$ – Crystal field interactions

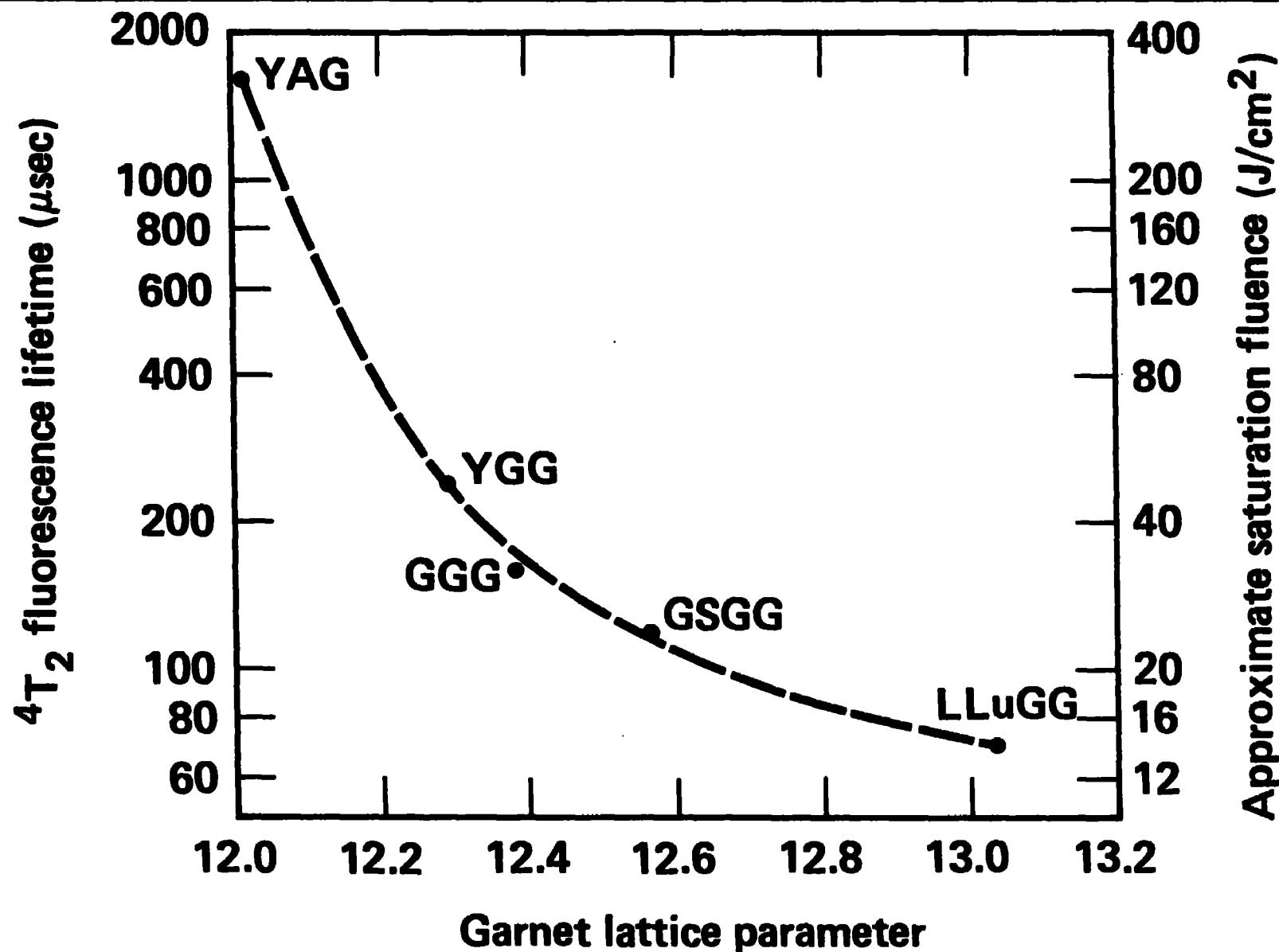


d^3 PHONON TERMINATED LASER – PROPERTIES

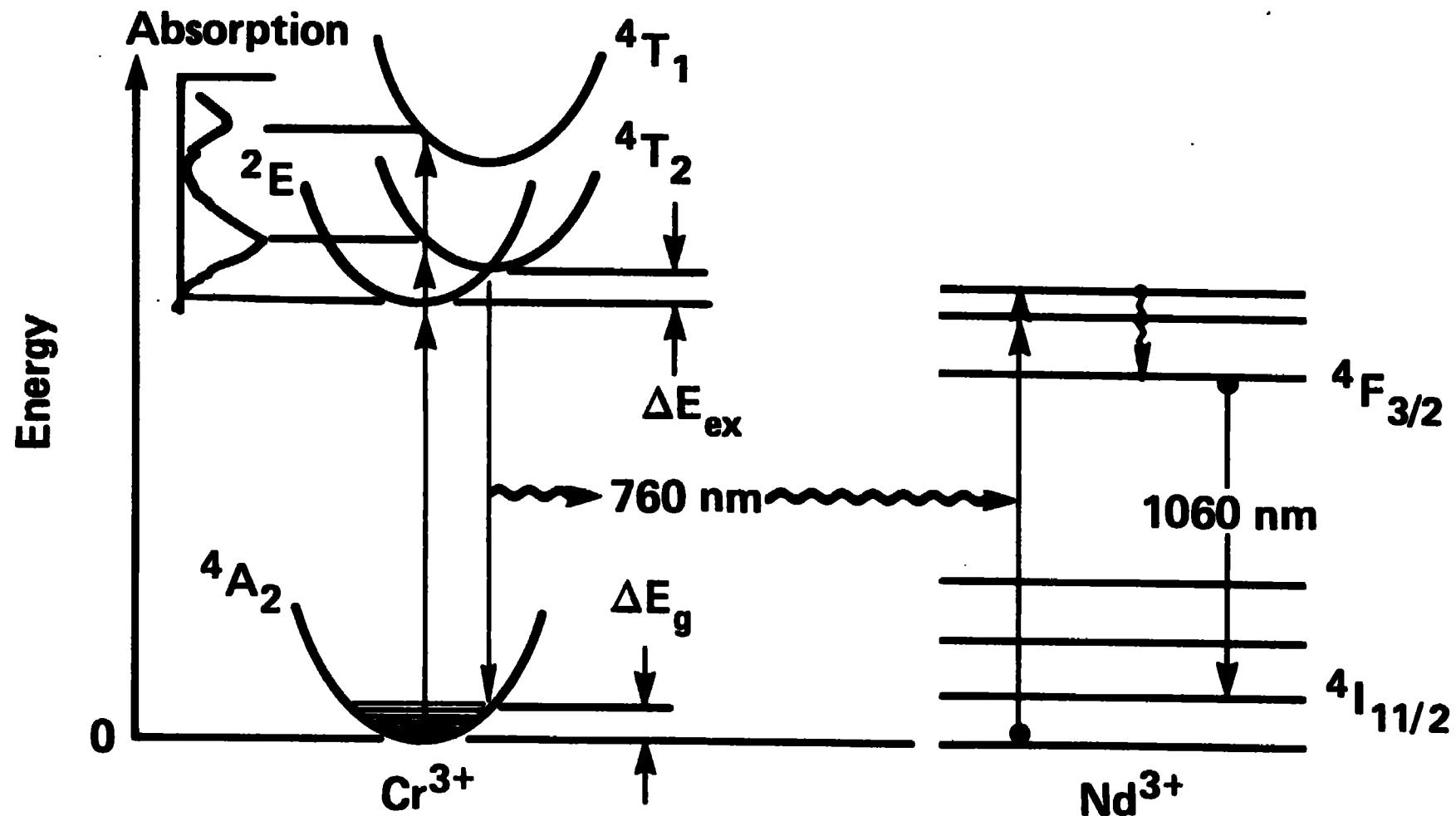


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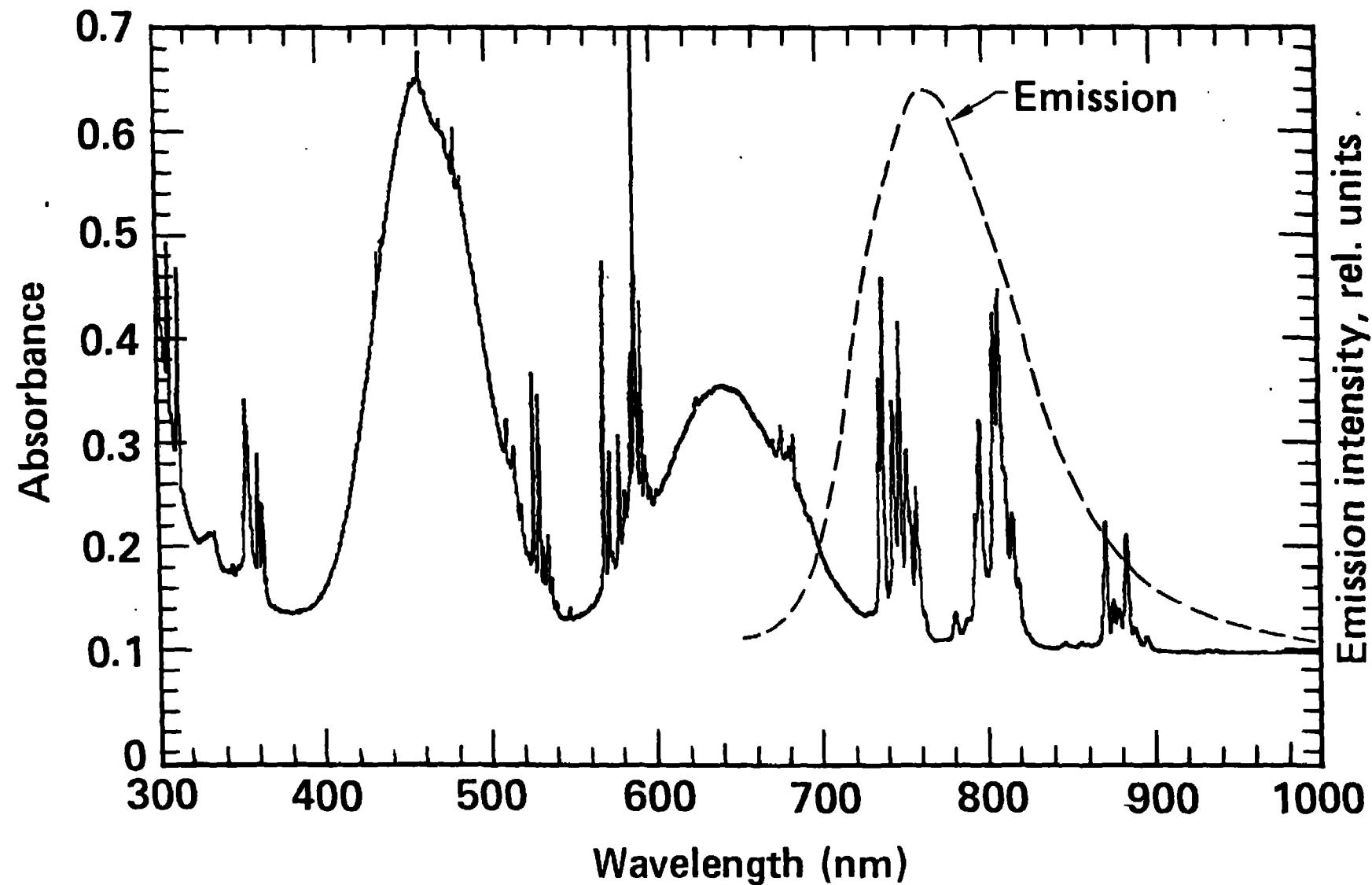
Systematics of Cr:Garnet 4T_2 fluorescence



$\text{Cr} \rightarrow \text{Nd}$ sensitization laser



Absorbance – Nd:Cr:GSGG

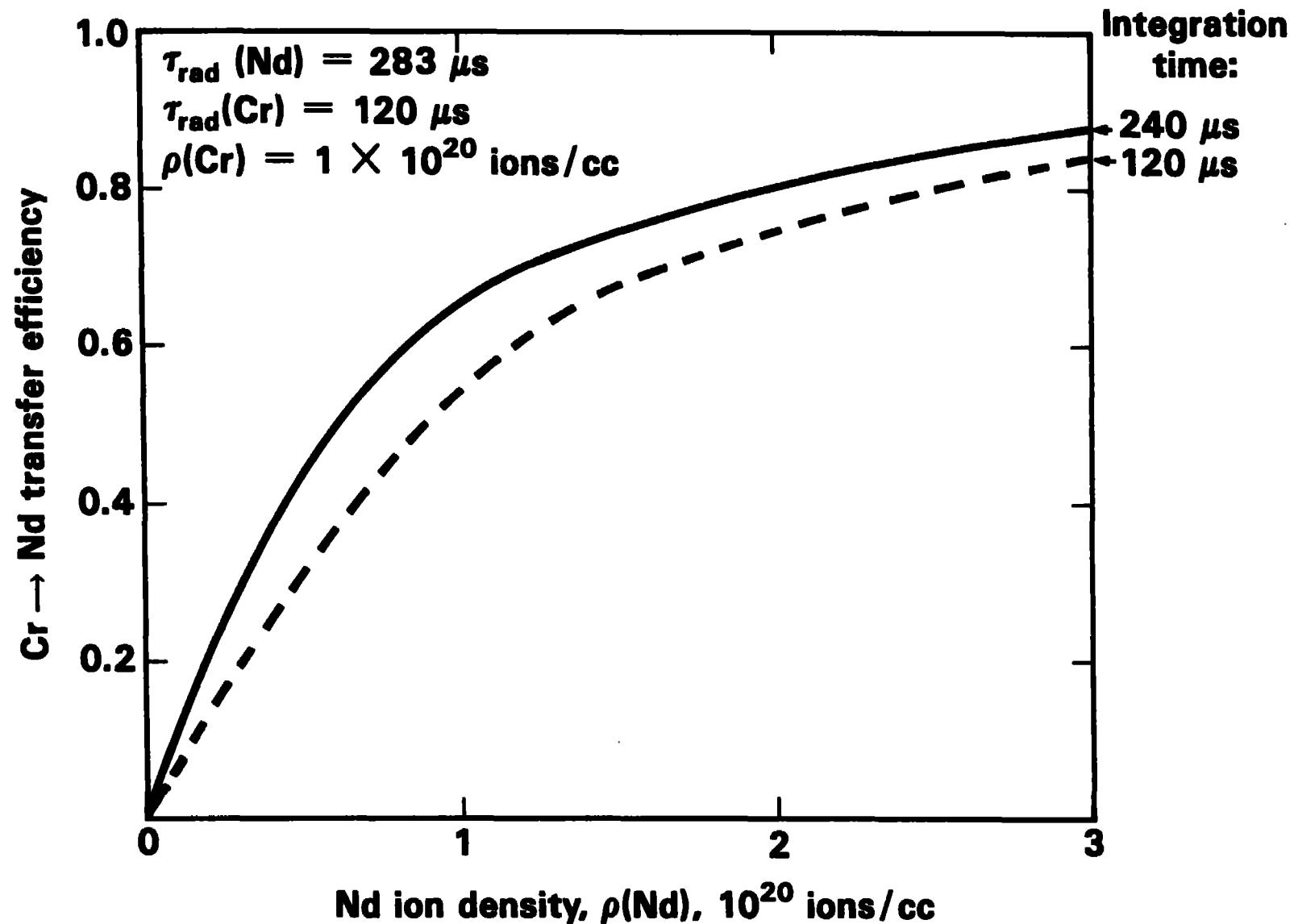


02-50-0684-1969

Cr → Nd transfer efficiency in GSGG



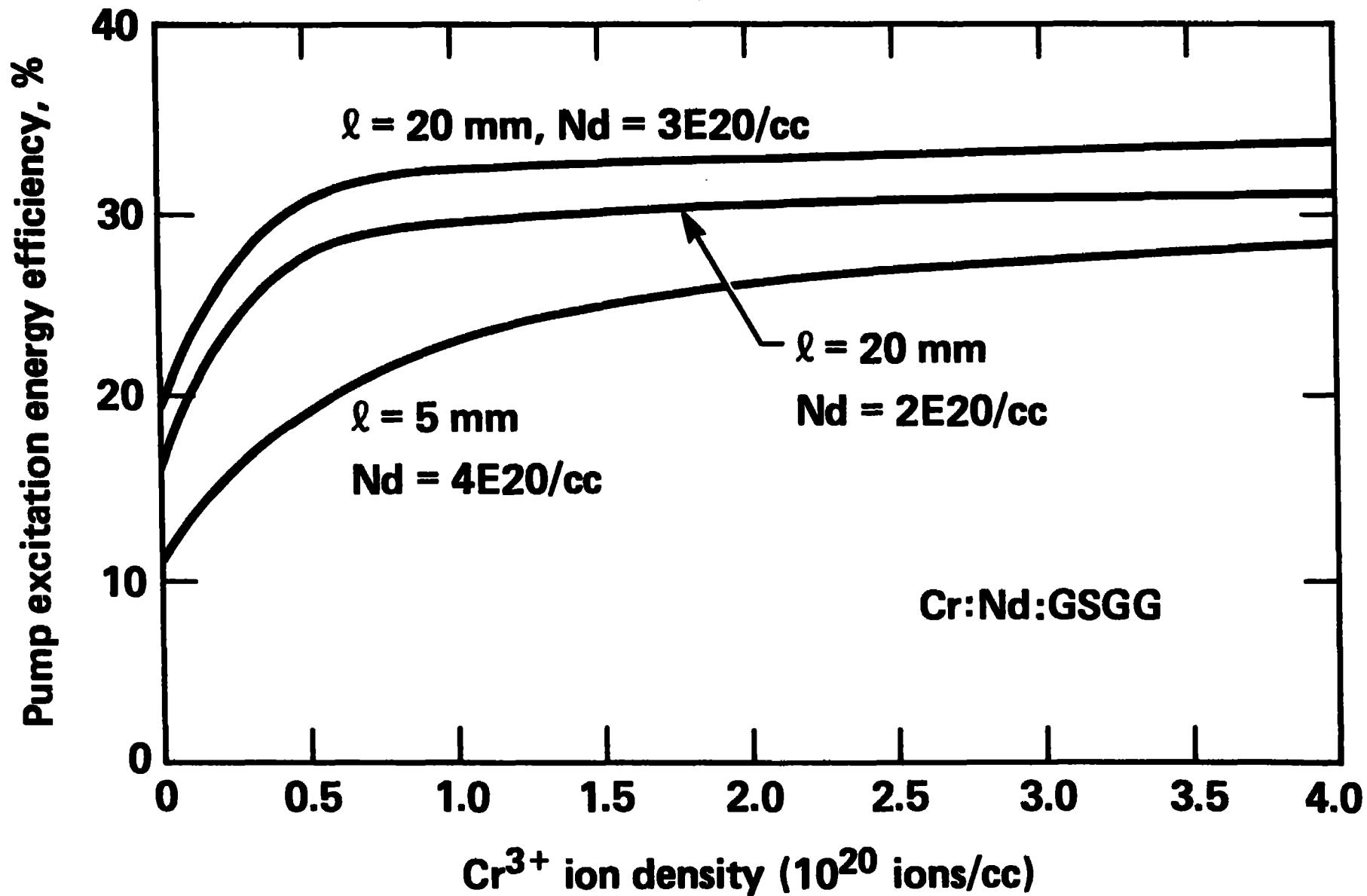
Data: Zharikov, et al. Sov. J.Q.E. 13, 82 (1983); 12, 338 (1982).



$^{4}F_{3/2}$ excitation energy efficiency (xenon flashlamp)



Lamp radiative efficiency = 75% $I = 1800 \text{ A/cm}^2$



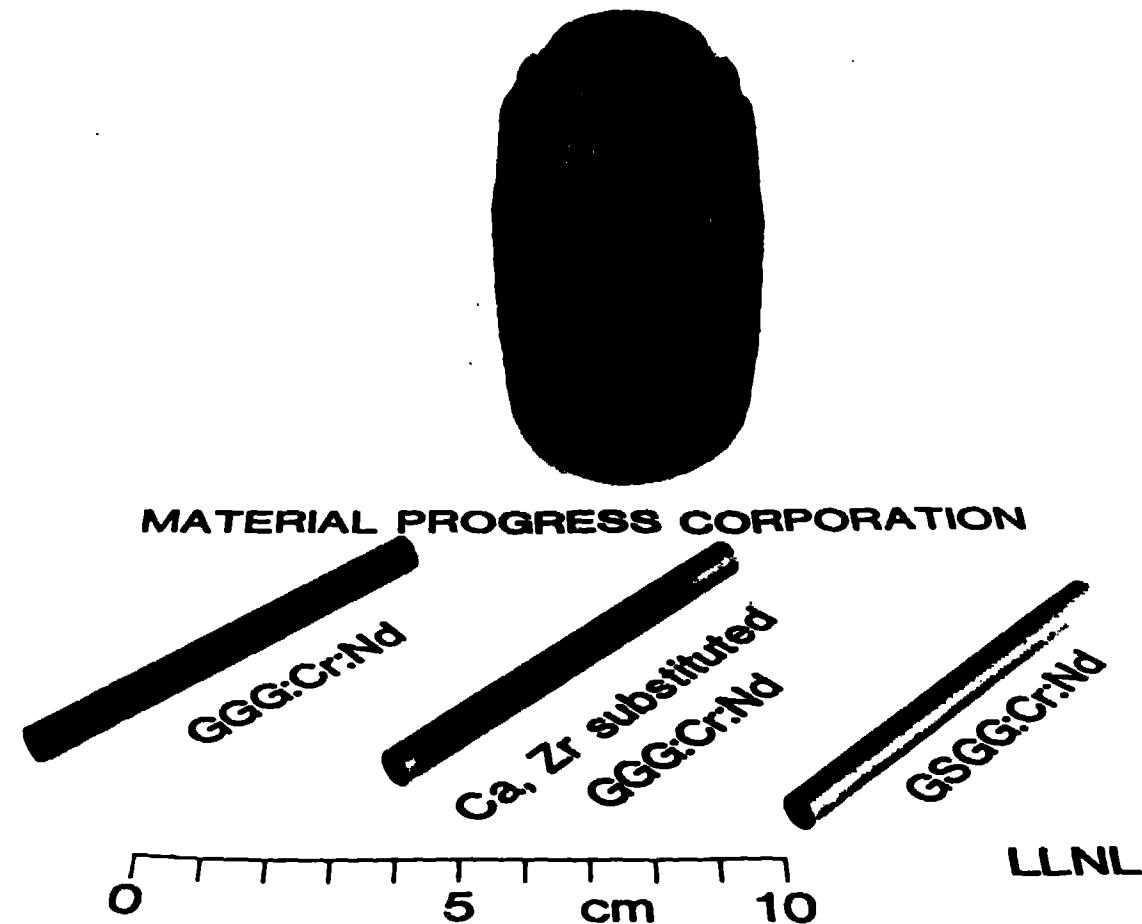
GGG and GSGG laser materials*



***Growth: F. Bruni (Material Progress Corp.)**

02-50-0684-1989

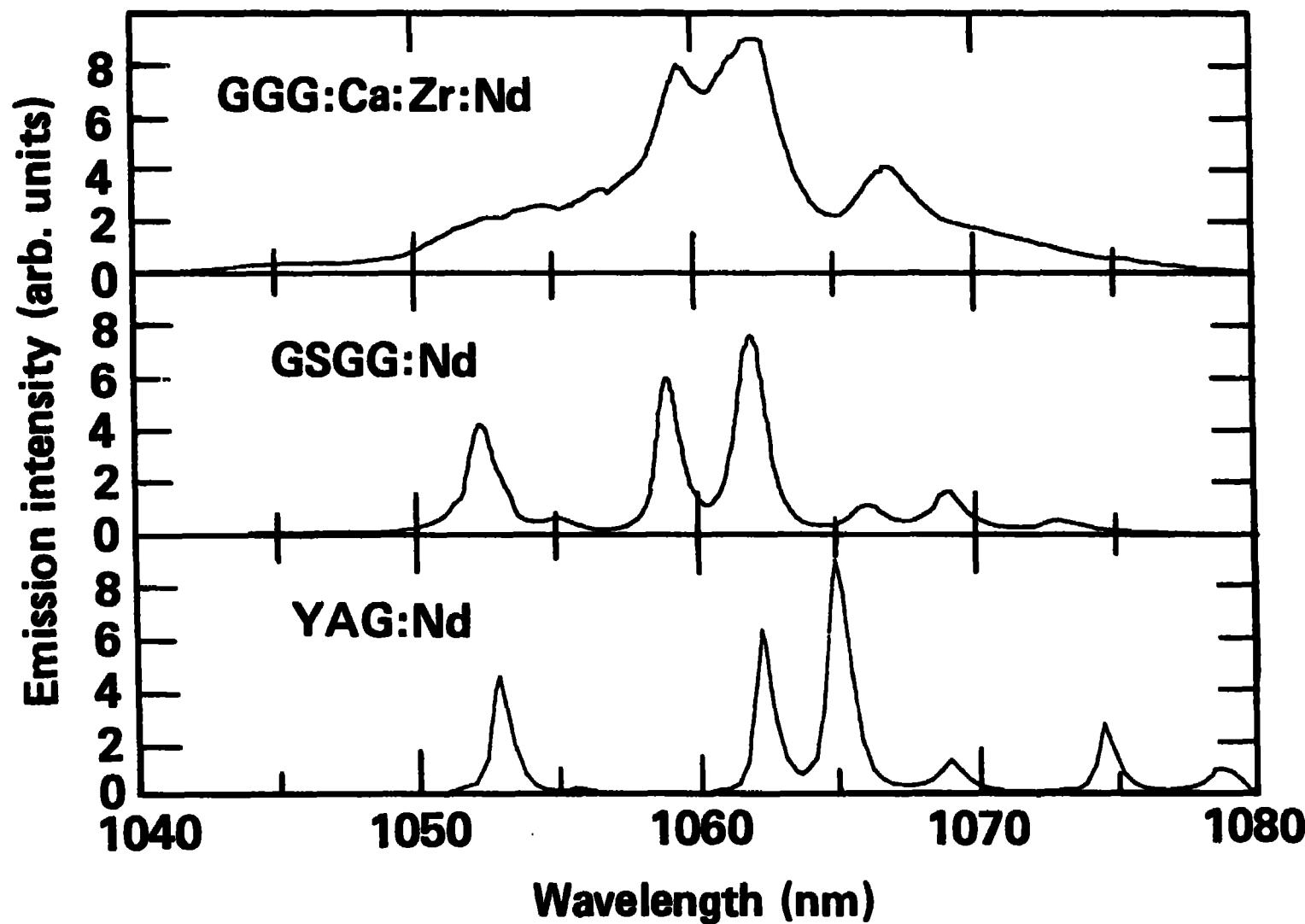
Oxide garnet laser materials*



*Growth: F. Bruni (Material Progress Corp.)

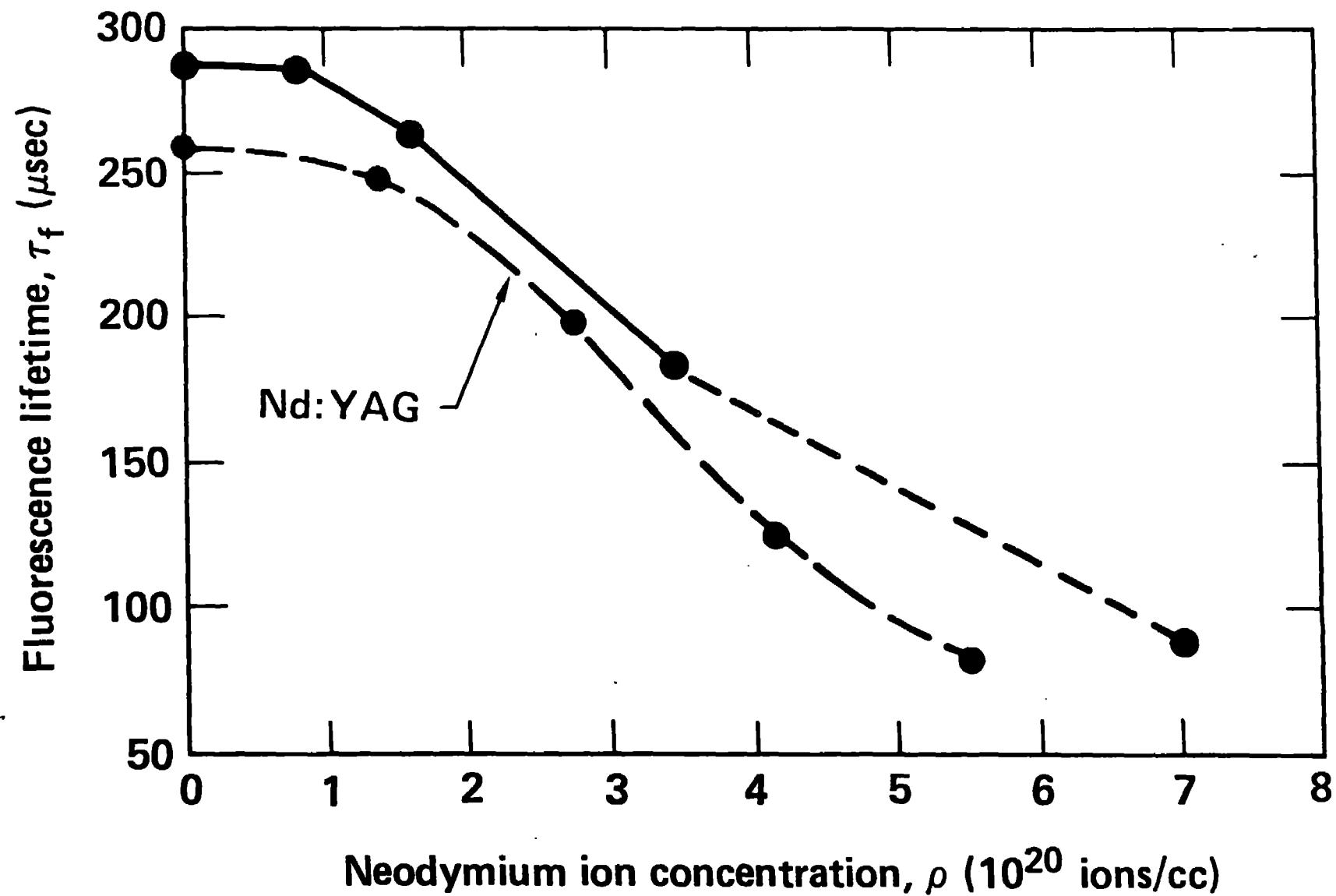
02-50-0684-1985

$^{4F}_{3/2} \rightarrow ^{4I}_{11/2}$ fluorescence – oxide garnets (300°K)



02-50-0684-1968

Nd:GSGG – fluorescence-concentration quenching



02-50-0684-1815

Spectroscopic laser parameters (I)



<u>Property</u>	<u>Nd:GSGG</u>	<u>Nd:YAG</u>
Judd-Ofelt param. Ω_2 , Ω_4 , Ω_6 (10^{-20} cm 2)	0.35, 2.35, 3.23	0.2, 2.7, 5.0
${}^4F_{3/2}$ - J.O. calc. radiative lifetime	283 μ sec	259 μ sec
Zero concentration lifetime	286 μ sec	\sim 260 μ sec
Half-lifetime quenching conc.	$\sim 5 \times 10^{20}$ ions/cc	$\sim 4 \times 10^{20}$ ions/cc
${}^4F_{3/2}$ Stark splitting	60 cm $^{-1}$	84 cm $^{-1}$
$R_2 \rightarrow Y_3$ peak wavelength	1061 nm	1064 nm
$R_2 \rightarrow Y_3$ peak cross-section, σ_L	3.2×10^{-19} cm 2	7×10^{-19} cm 2
Effective small-signal gain cross section [†] , σ_{eff}	1.4×10^{-19} cm 2	2.8×10^{-19} cm 2
Homogeneous saturation fluence*	1.0 J/cm 2	0.5 ₀ J/cm 2

*Does not account for orientational hole-burning.

[†] $\alpha = \sigma_{eff} \Delta N({}^4F_{3/2})$

Spectroscopic laser parameters (II)



<u>Property</u>	<u>Cr:GSGG</u>
$^4A_2 \rightarrow ^4T_2$ peak wavelength	640 nm
$^4A_2 \rightarrow ^4T_2$ peak cross-section, σ_A	$3.3 \times 10^{-20} \text{ cm}^2$
$^4T_2 \rightarrow ^4A_2$ peak cross-section, σ_E	$0.8 \times 10^{-20} \text{ cm}^2$
4T_2 fluorescence lifetime	120 μsec
Half-lifetime quenching conc.	$> 3 \times 10^{20}$ ions/cc

GSGG material properties (preliminary)



<u>Property</u>	<u>Cr:Nd:GSGG</u>	<u>Nd:YAG</u>
Thermal conductivity, κ	7-9 W/m°C (conc. dep.)	~13 W/m°C
Thermal expansion, α	$7.8 \times 10^{-6}/^{\circ}\text{C}$	$7.8 \times 10^{-6}/^{\circ}\text{C}$
Young's modulus, E	~21,000 kg/mm ²	~31,700 kg/mm ²
Breaking strength [†] (4-pt bend), S _T	15-20 kg/mm ²	~20 kg/mm ²
Poisson's ratio, ν	0.11-0.18	Not reported
Thermal shock*, R _T	~700 W/m	~800 W/m
Index of refraction, 1060 nm	1.925	1.815
Density	6.46	4.55
Nd-segregation coeff.	0.65 ± 0.04	0.18
Cr-segregation coeff.	1.0 ⁻	—
10 ns damage threshold	$\begin{cases} 16 \text{ J/cm}^2, \text{ surface} \\ >28 \text{ J/cm}^2, \text{ bulk} \end{cases}$	

$$*R_T = (1 - \nu) \kappa S_T / \alpha E$$

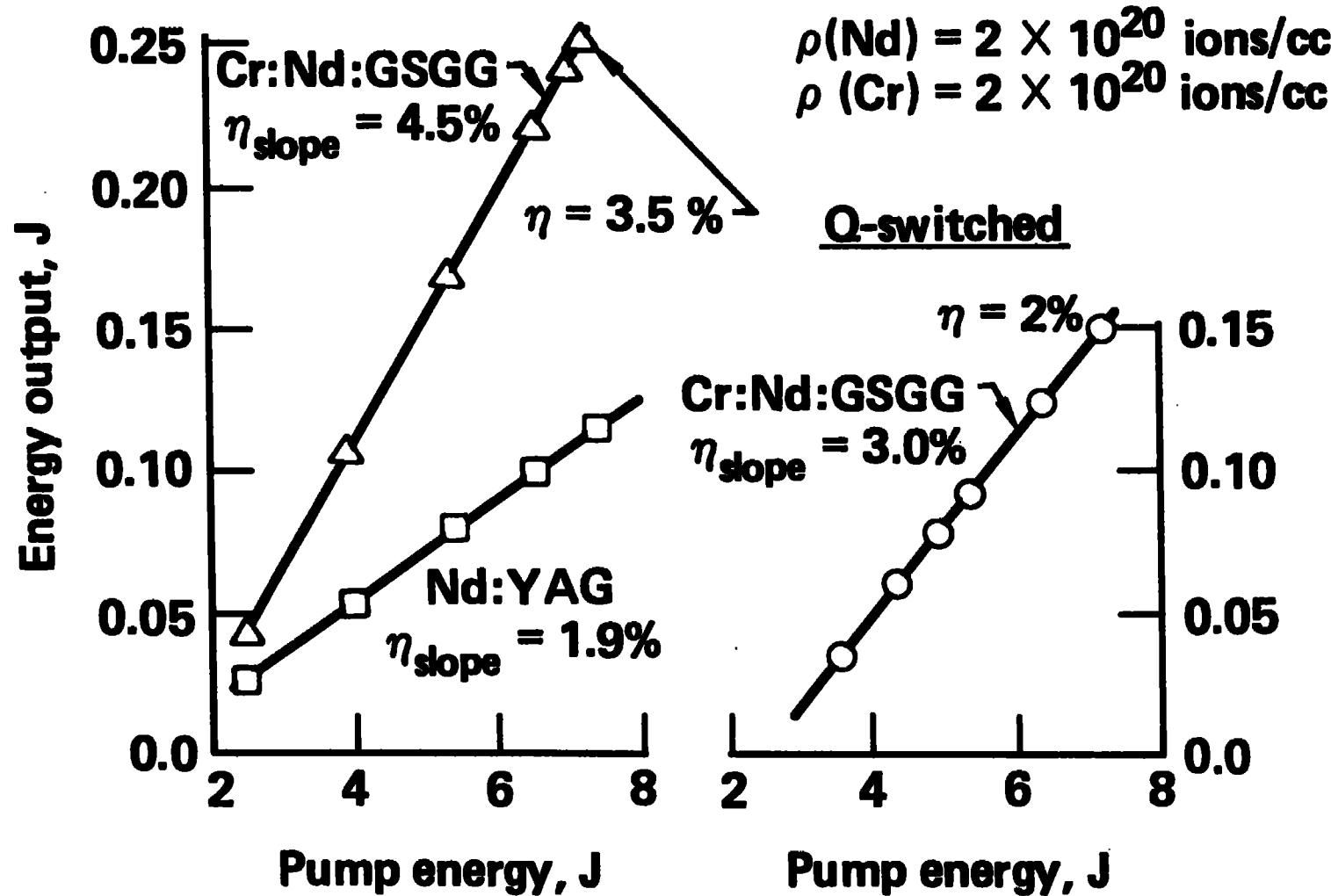
[†]Depends on fabrication

Laser performance* – Cr:Nd:GSGG



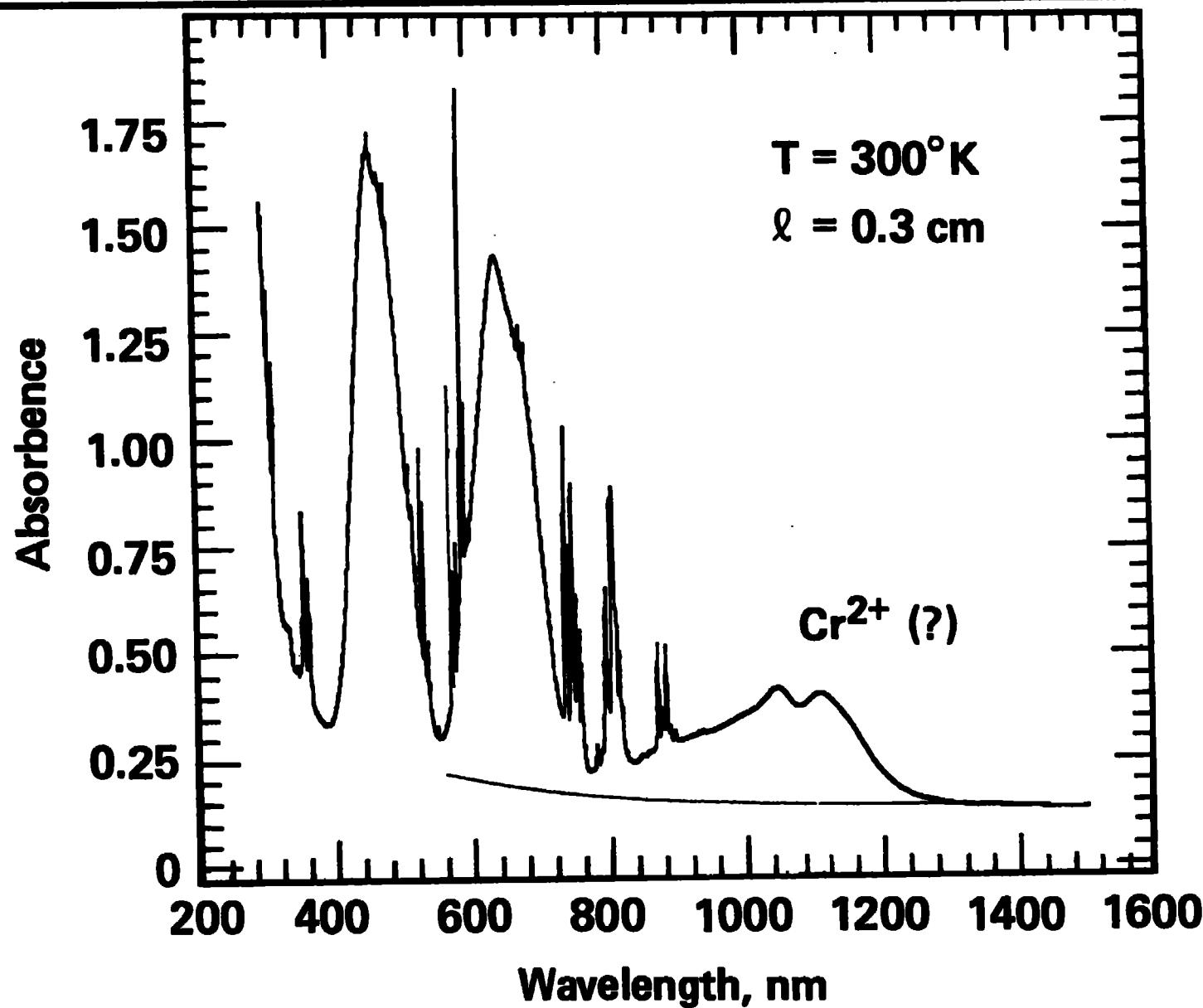
Long-pulse ($R = 0.66$)

$\phi = 5 \text{ mm}$ $I = 50 \text{ mm}$



*E.V. Zharikov, et al., Sov. J. Quantum Electron., 13, 1306 (1983).

1000 nm loss in selected Cr:Nd:GSGG crystals



Tunable chromium-doped laser materials

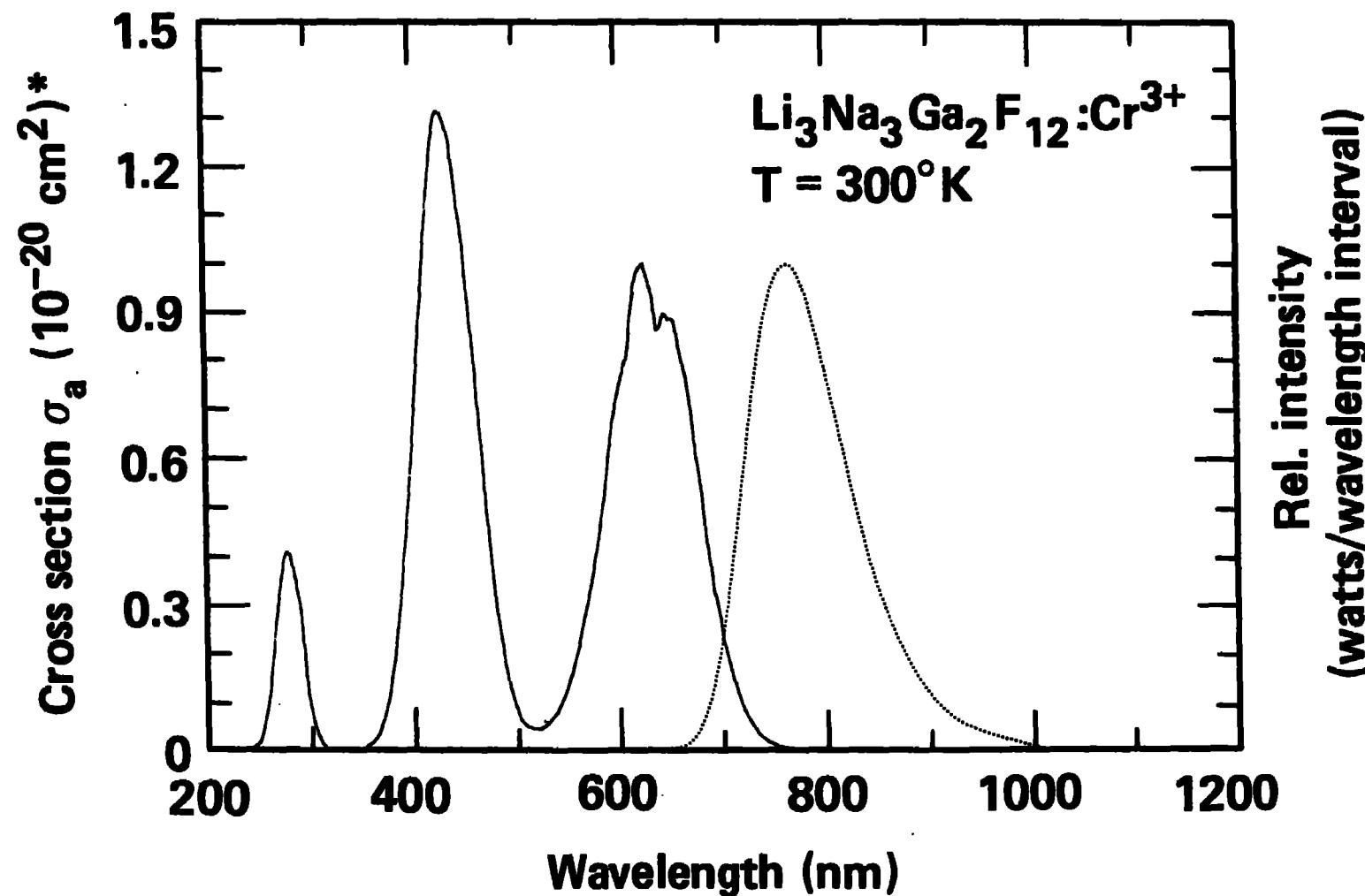


<u>Crystal Type</u>	<u>Composition</u>	<u>Peak Fluor.-λ (nm)</u>	<u>Pump Sources</u>
Oxide garnet	$\text{Gd}_3\text{Ga}_2\text{Ga}_3\text{O}_{12}$	760	Kr
Oxide garnet	$\text{Gd}_3\text{Sc}_2\text{Ga}_3\text{O}_{12}$	770	Kr; flashlamp
Oxide garnet	$\text{Y}_3\text{Ga}_2\text{Ga}_3\text{O}_{12}$	740	Kr
Oxide garnet	$\text{Y}_3\text{Sc}_2\text{Ga}_3\text{O}_{12}$	760	Kr
Crysoberyl	BeAl_2O_4	750	Kr; flashlamp; Hg
Emerald	$\text{Be}_3\text{Al}_2(\text{SiO}_3)_6$	750	2 X Nd:YAG + SRS; flashlamp
Scheelite	ZnWO_4	990	Kr
Fluoride perovskite	KZnF_3	790	Kr
Fluoride garnet	$\text{Li}_3\text{Na}_3\text{Ga}_2\text{F}_{12}$	765	Pulsed-dye

Spectral properties of fluoride garnet



$$*\rho(\text{Cr}) = 2.1 \times 10^{20} \text{ ions/cc}$$



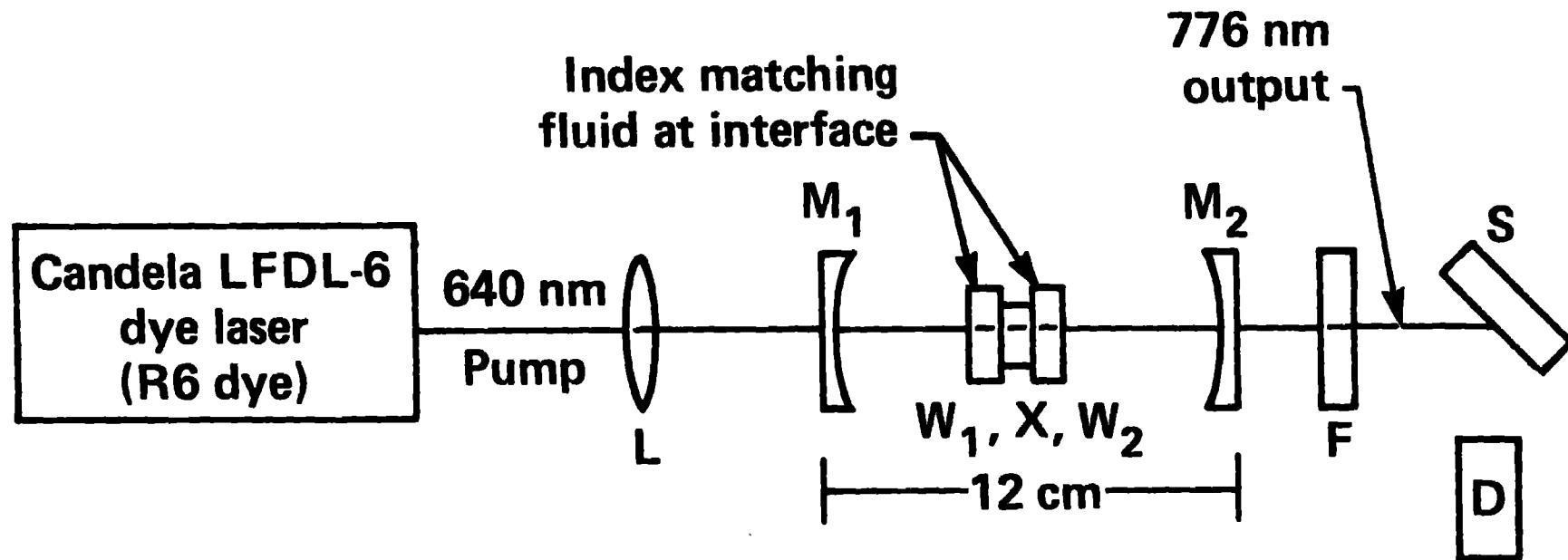
Chromium doped fluoride garnet laser*



***Growth: H. Guggenheim (AT&T - Bell Labs)**

02-50-0684-1988

Set-up for fluoride garnet laser experiment



X $\text{Li}_3\text{Na}_3\text{Ga}_2\text{F}_{12}:\text{Cr}$ laser crystal (0.5 cm thick)

W₁,W₂ Fused silica windows, AR coated outside

M₁,M₂ 25 cm radius mirrors, max R 750-800 nm

L 25 cm focal length lens

F Long wavelength pass filter

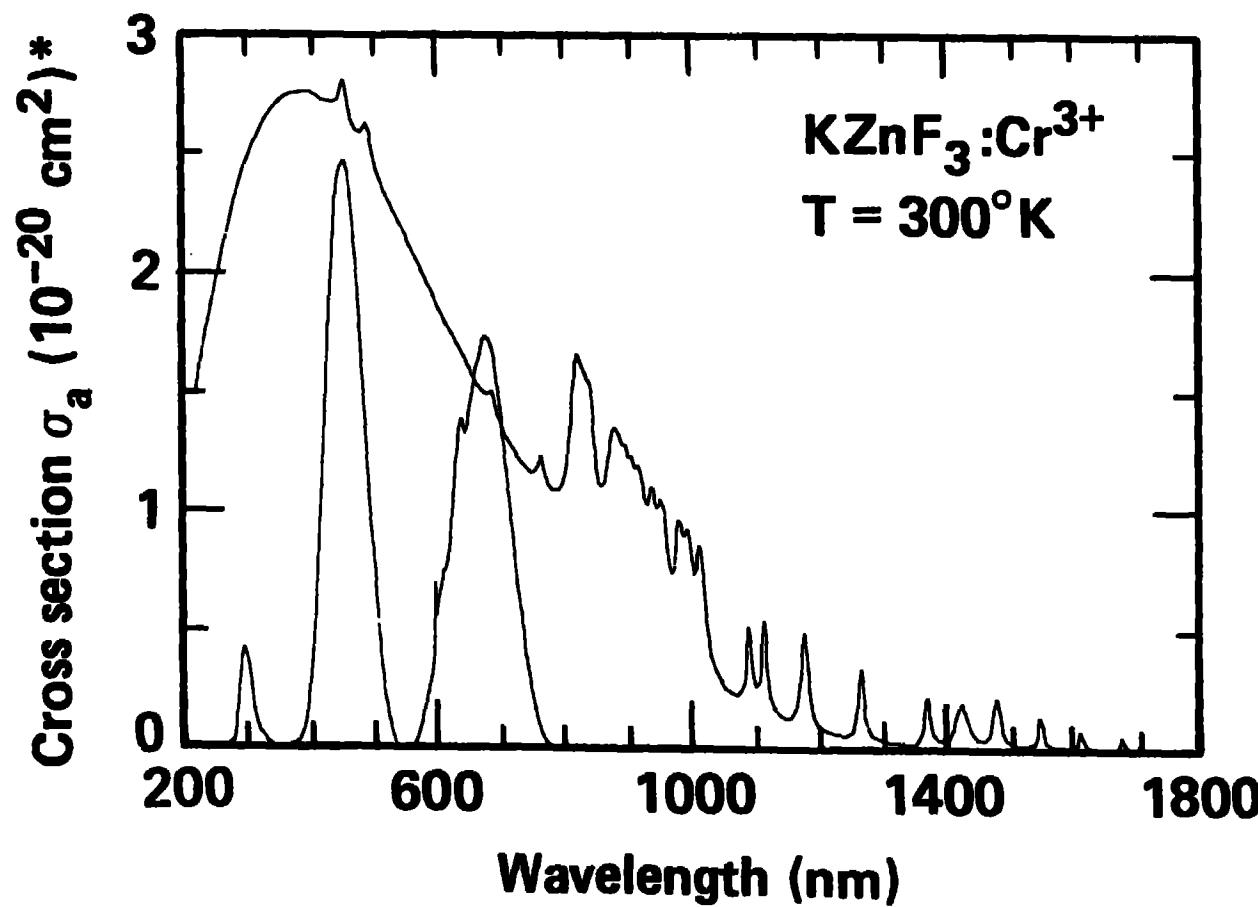
S Diffuse scatterer

D Photodiode detector

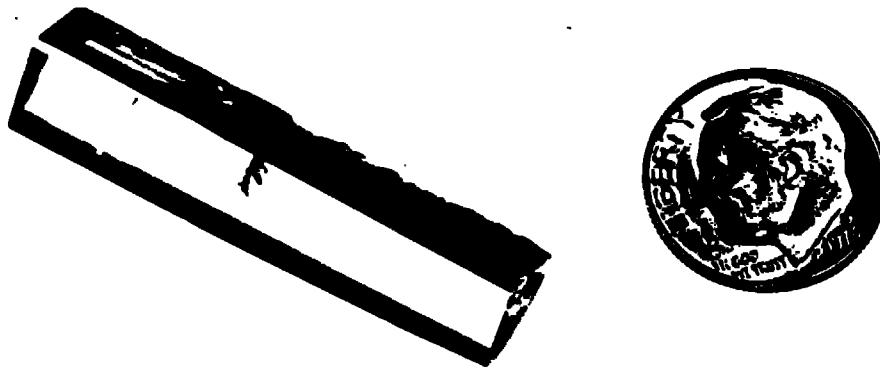
Xenon lamp – fluoride perovskite coupling



* ρ (Cr) = 3.7×10^{19} ions/cc



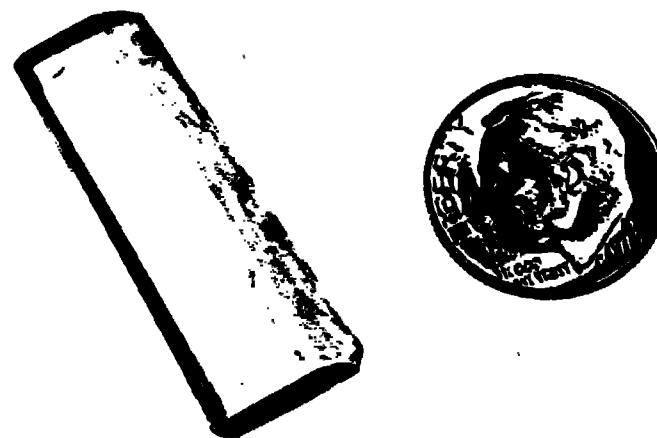
Chromium doped fluoride perovskite laser*



***Growth: H. Newkirk (LLNL)**

02-50-0684-1991

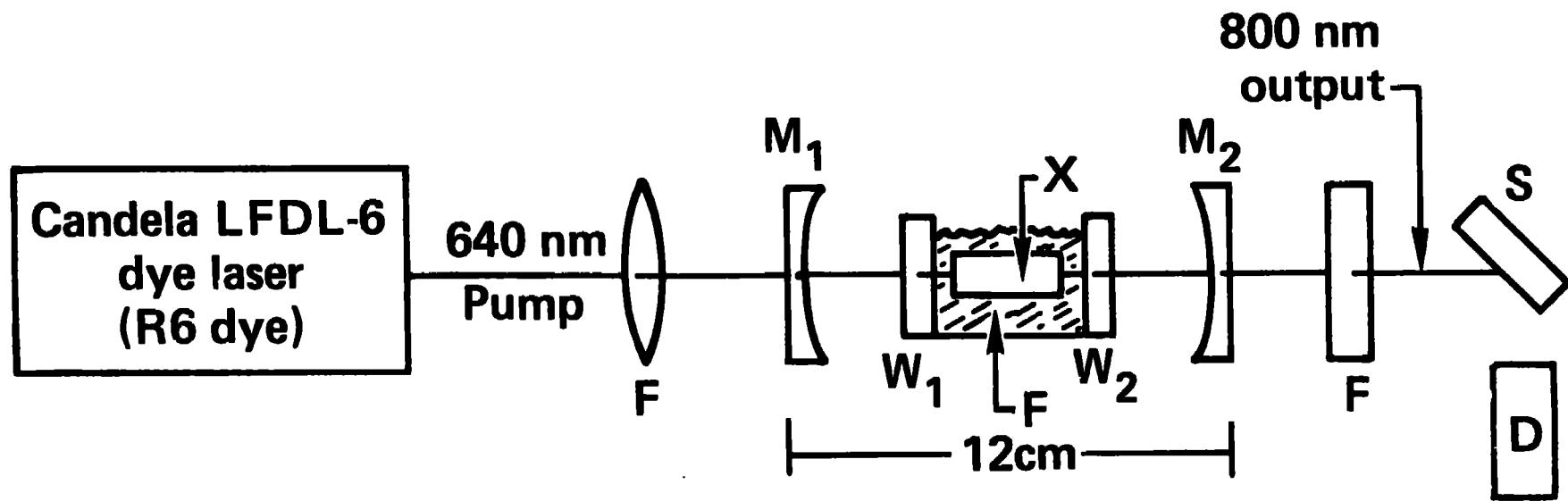
Chromium doped fluoride perovskite laser*



*Growth: H. Guggenheim (AT&T - Bell Labs)

02-50-0684-1987

Set-up for fluoride perovskite laser experiment



X KZnF₃:Cr laser crystal (4 cm long)

F Index matching fluid

W₁,W₂ Fused silica windows, AR coated outside

M₁,M₂ 25 cm radius mirrors, max R 750-800 nm

L 25 cm focal length lens

F Long wavelength pass filter

S Diffuse scatterer

D Photodiode detector

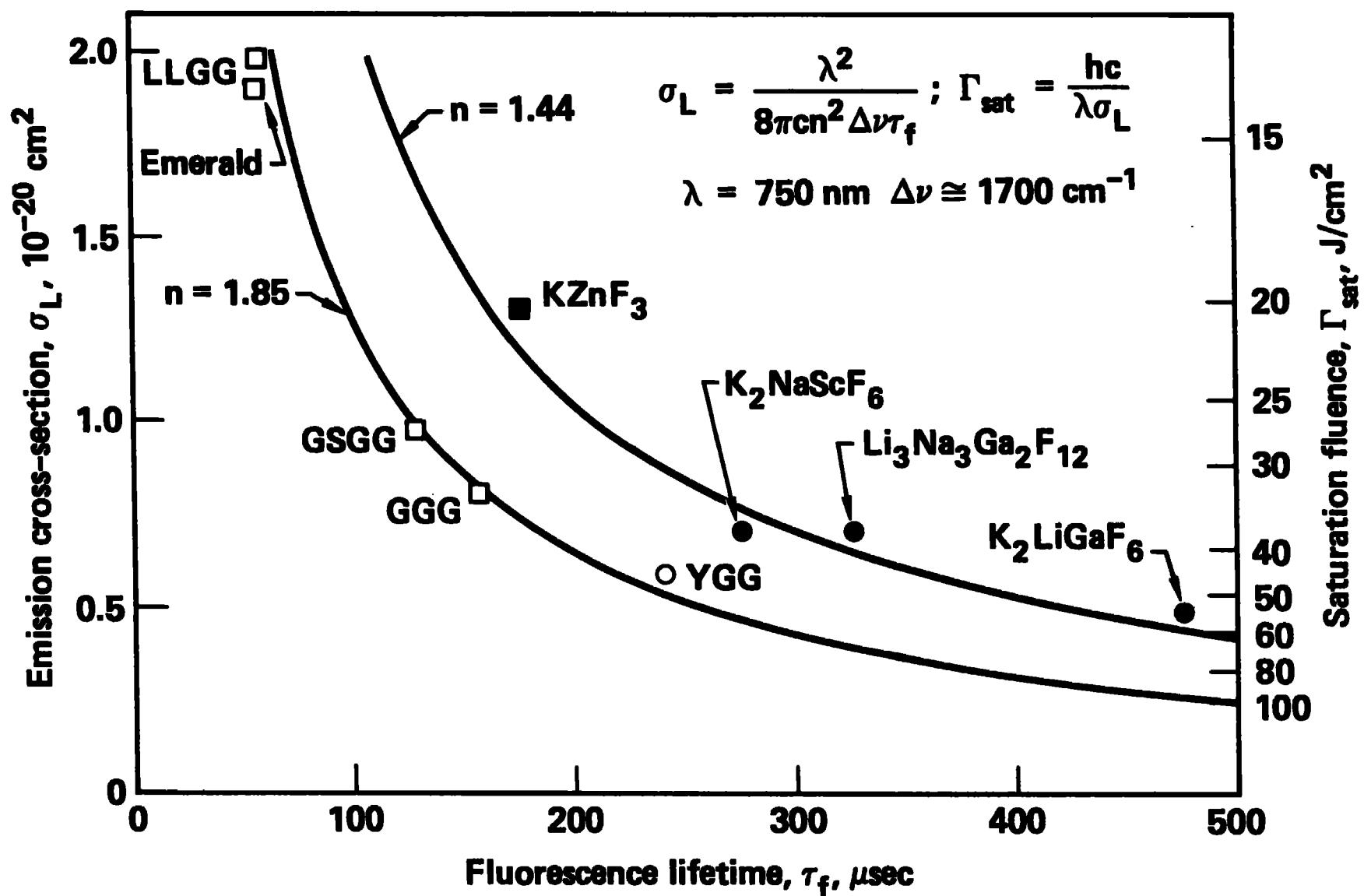
Laser parameters for Cr³⁺ — gain media



Crystal	Name	n	τ_f (μs) 300°K	$\tilde{\Delta\nu}$ (cm ⁻¹)	λ_p (nm)	σ_p (pm) 300°K	$\Gamma_{\text{peak}}^{\text{sat}}$ (J/cm ²)
BeAl ₂ O ₄	Alexandrite	1.74	240	~1800	750	0.6	~44
Be ₃ Al ₂ (SiO ₃) ₆	Emerald	1.75	60	~2000	725	1.9	~14
Gd ₃ Ga ₅ O ₁₂	GGG	1.94	160	~1600	740	0.8	~33
Gd ₃ Sc ₂ Ga ₃ O ₁₂	GSGG	1.90	120	~1700	760	1.0	~27
Y ₃ Ga ₅ O ₁₂	YGG	1.96	240	~1300	740	0.6	~45
Y ₃ Sc ₂ Ga ₃ O ₁₂	YSGG	1.86	(140)	~1700	760	0.9	~28
La ₃ Lu ₂ Ga ₃ O ₁₂	LLGG	(1.95)	70	~1750	820	1.9	~13
K ₂ NaScF ₆	Elpasolite	1.44	280	~2000	775	0.7	~37
K ₂ LiGaF ₆			480	~1660	755	0.5	~57
Li ₃ Na ₃ Ga ₂ F ₁₂	Garnet	1.43	330	~1600	760	0.7	~37
KZnF ₃	Perovskite	1.46	176 85*	~1700 ~2000	795 800	1.3 10.0*	~19 ~2.5*

*Brauch and Dürr, Opt. Comm. 49, 61 (1984); inconsistent with $\sigma_p = \lambda_p^2 / 8\pi c n^2 \tilde{\Delta\nu} \tau_f$

Parameters of Cr³⁺ (3d³) laser gain media



Developments expected in '85



Co-doped (Cr:Nd) materials

- Scaled-growth of GSGG – $10 \times 20 \text{ cm}^2$ slabs
- Increased efficiency $> 6\%$
- Evaluation of alternative oxide garnets
 - $\text{Gd}(\text{GaCaZr})\text{GaG}$
 - GdInGaG
 - GdScAlG

Chromium-doped tunable materials

- Materials with larger stimulated emission cross-sections ($\sigma \geq 2 \times 10^{-20}$; $\Gamma_{\text{sat}} < 15 \text{ J/cm}^2$)